

PROGRESS REPORT ON WORKS CONCERNING
SILICA AEROGELS

M. Cantin

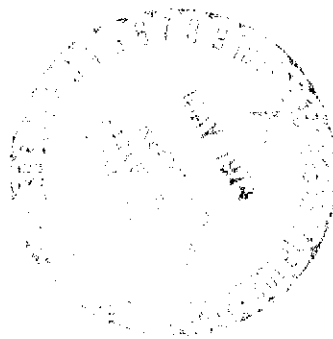
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Introduction

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The production of light by the Cerenkov effect is a function of the refraction index of the environment. The number of photons produced is given by

$$\frac{dN}{dl} = k \left(\lambda - \frac{\lambda}{\beta^2 n^2} \right)$$

where k = a constant;

β = the ratio of the particle velocity to the velocity of light in a vacuum;

dl = unit of length.

Messrs. Linney and Peters¹ have shown that porous silica can, under certain conditions, have a refraction index included between the indices for air and silica with the value of the index being a function of the percentage of volume occupied by each of the two substances, air and water. This is on condition that the silica particles are quite small relative to the wavelength of light.

Starting from this principle we thought that silica aerogels could provide an ingenious solution to this problem, in particular when the Teichner and Nicolaon preparation method² was used.

This report provides some results concerning the use of silica aerogels as a Cerenkov converter.

¹Nucl. Inst. and Meth. 100 (1972) 545.

²Patent no. 1,568,817.

*Numbers in margin indicate foreign pagination.

1. Silica Aerogels

This study was performed at Lyon, at the Faculty of Sciences, in the laboratory of Teichner, under the direction of Gardes, according to the works of Nicolaon (cf. the Mission Report of Cantin, Casse and Koch SEP/SES/73.7 of 19 January 1973).

We took as a starting base the values given by Nicolaon. To a mixture of 3 cc orthosilicate, 1.2 cc water, and 0.3 cc ammonia, we add 7 cc methanol in order to produce a 40% mixture; 3 cc for a 50% mixture, 2 cc for a 60% mixture, etc... These mixtures put in the autoclave have allowed us to plot the curve of the density of aerogel as a function of the percentage of orthosilicate.

Under these conditions, the aerogel is transparent for a percentage of orthosilicate less than 40%. Above this value, the aerogel becomes opaque, eventually entirely so with 70% orthosilicate.

This opacification has been reduced to zero by reducing the volume of ammonia in the reaction.

The volume of water was reduced to the maximum below a certain volume. The mixture then became viscous and the bubbles trapped during the mixture no longer came together to escape.

For a percentage of orthosilicate greater than 80%, water and orthosilicate are no longer miscible. A pellicle of gel is formed at the junction of the two liquids. This pellicle isolates both liquids.

The reaction giving the silica gel is exothermic. When the percentage of orthosilicate reaches and exceeds 60%, the quantity of heat is released in a reduced volume and is not removed, thus accelerating the reaction. The increase in heat produces stresses which break the gel. The study of the various parameters have shown us that the time required for gelification is inversely proportional to the percentage of methanol. In order to avoid breaks in the gel and at the same time producing a dense and transparent solid, we reduced the volume of ammonia. The breaking of the gel increases proportionally as the volume becomes greater.

Nevertheless, it appears that the decrease in volume of the ammonia inhibits breaking of the gel and improves the transparency. However, this produces a more fissured aerogel.

The last results were obtained with 20 cc of CH_3OH , 18 cc of H_2O , .1 cc of NH_3 at 1.25% added to 72 cc orthosilicate producing a density of 0.33. 13

The gel was dried in an autoclave. The time required to reach critical temperature (260°C) and the time for removal of gases are important parameters. With a rise in 1h 30 min and a removal in 15 min, the gel is found to be broken into 0.25 cm^3 fragments. With a rise in 4 hours and a removal in 30 min, the fragments have mean sizes of 1 cm^3 .

We sought to increase the density of the aerogel by adding silica powder to the mixture before gelification. The resulting mixture was totally opaque with a maximum density of 0.42. Such a method appears to be completely inadequate given the great opaqueness of the resultant aerogel.

In conclusion, a more detailed study should allow improvement in transparency and avoid fracturing the aerogel. We believe that this method cannot produce aerogels with densities greater than 0.4.

The present autoclave allows, at maximum, production of a cylinder measuring 6 cm in diameter and 20 cm in height.

We have briefly covered the problems. There are many of them but there are an infinite number of parameter combinations possible. The qualities of the aerogel are functions:

- of properties of the mixture: water, methanol, ammonia and orthosilicate;
- of the temperature at which the mixture is made;
- of the time taken by the autoclave to reach the critical temperature;
- of the time taken to remove gaseous methanol from the autoclave;
- of the autoclave cooling time.

II. Transparency Qualities of Silica Aerogels

The photons produced by the Cerenkov effect in the converting medium can be observed by photomultipliers and, for this reason, the converting material

should not absorb them. Knowing that the number of photons produced is proportional to $1/\lambda^2$, it is advantageous to have a good transparency in the short wavelengths.

Figures 1 and 2 provide the transmission coefficient of a silica aerogel with a density of 0.16 and having a 4 mm thickness for a wavelength of from 2,600 to 25,000 Å. The absorption peaks in the infrared are characteristic of the material. Nevertheless, absorption peaks are noted at 11,700 and 16,800 Å which is not the case with the silica powder manufactured by the firm Aérosil (Figure 3).

There may be seen a decrease in transmission factor in the visible and ultraviolet part of the spectrum. This is caused by a scattering of the low wavelength light. Since we use these converters in a scattering assembly, this phenomenon is not harmful.

The few measurements made show us that the absorption of light is more connected with the conditions of manufacture (quantity of NH_3) than with the density of the product.

We hope to make in the near future absorption measurements of light as a function of wavelength.

For the time being, only gross measurements have been made at the same time using the Cerenkov effect in the material. When we assume that 3 mm of quartz have a zero coefficient of absorption, we find for the aerogels:

$$A_1 = (1 - 0.94) \text{ for } 25 \text{ mm};$$

$$A_2 = (1 - 0.55) \text{ for } 37 \text{ mm};$$

$$A_3 = (1 - 0.285) \text{ for } 33 \text{ mm}.$$

A simple observation of samples confirms these figures.

Progress made in manufacturing technology shows us a constant improvement in transparency of the material.

A coarse measurement of scattering was done with an aerogel. The findings are shown in Figure 4. We see that the photons pass preferentially over the top but a good proportion exit laterally.

III. Measurement of the Cerenkov Effect in Silica Aerogels

Since the purpose of the material is to be used as a converter in a detector operating on the Cerenkov effect principle, the measurement of the Cerenkov effect, while at the same time providing us with gross results, is a very important measurement.

1. Detector Used

This detector includes a scattering chamber 7.7 cm in diameter and 6.8 cm in height observed by a photomultiplier (PM) EMI 9708. Such an assembly has a scattering surface of 228 cm^2 for a photocathode surface of 30 cm^2 .

This assembly with a reduced volume is quite flexible and can be used for experimentation as an accelerator.

The disadvantage of such an assembly is the fact that the aerogel (about 70 cm^3) occupies a considerable volume in the scattering chamber (320 cm^3) and that the volume of aerogel disturbs the measurement insofar as it is not perfectly transparent.

2. Findings

We made tests as an accelerator using the scattering detector with one photomultiplier (PM) and 4 samples of aerogel. We have plotted in Figure 6 the response of the detector as a function of $1/\beta^2$ in which β is the ratio of the velocity of the particle to the velocity of light. The response is roughly as expected. Given the small number of measurement points, the value found for the index is only an indicative value.

Conclusions

These measurements involve small-sized samples. At the present time, our effort involves the manufacture of large-volume units.

This material is quite brittle owing to its great porosity. The next step should be to consider how to improve its mechanical service life. Here, we believe that sintering can be one solution.

We believe that an absorption of water can modify the characteristics of the material. For the time being, we are satisfied to keep the material

in it and the humidity. It is perhaps possible to cause the retraction index to vary by changing the humidity.

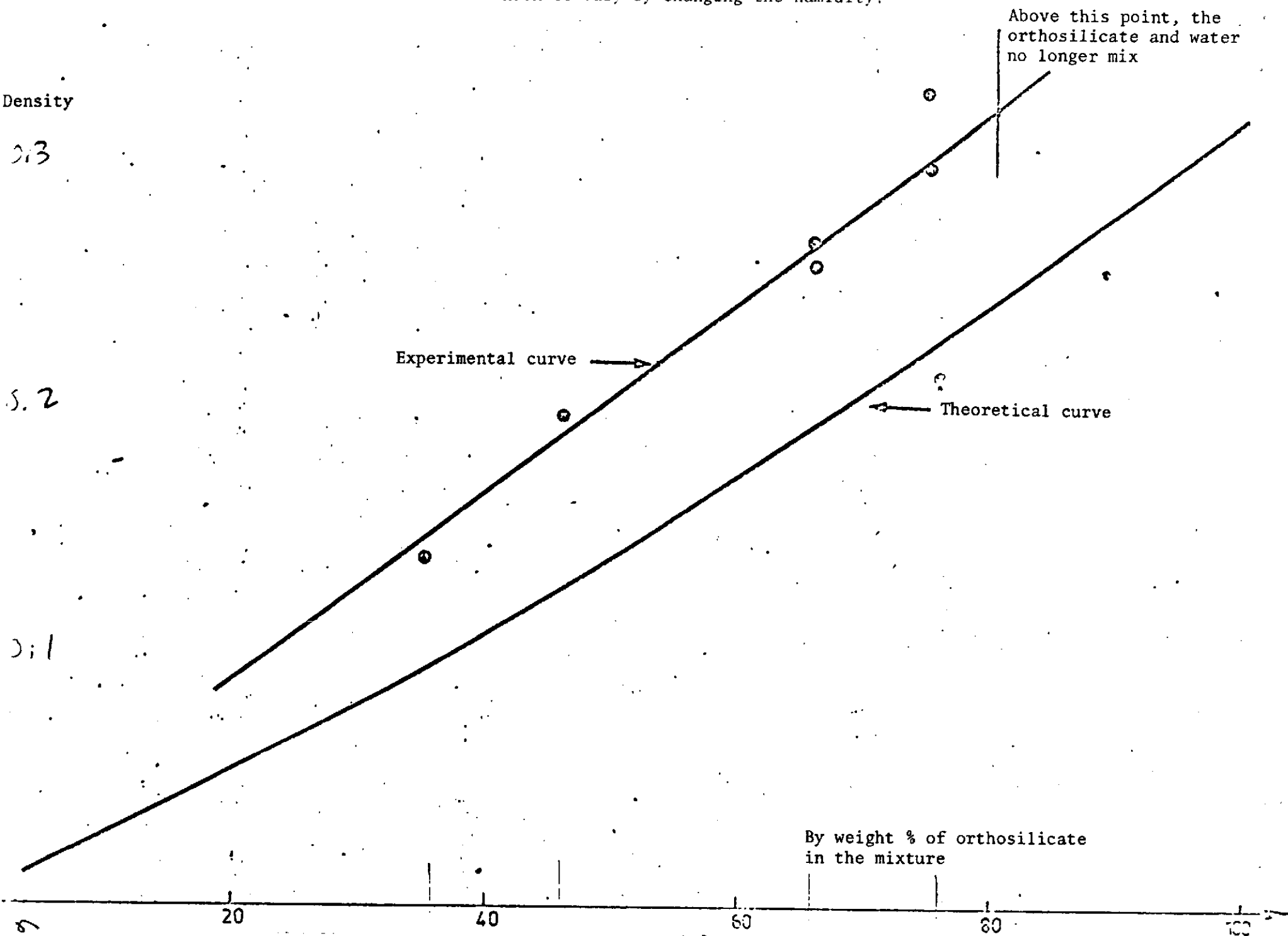


Figure 1. Density of the Silica Aerogel as a Function of the Percentage of Orthosilicate.

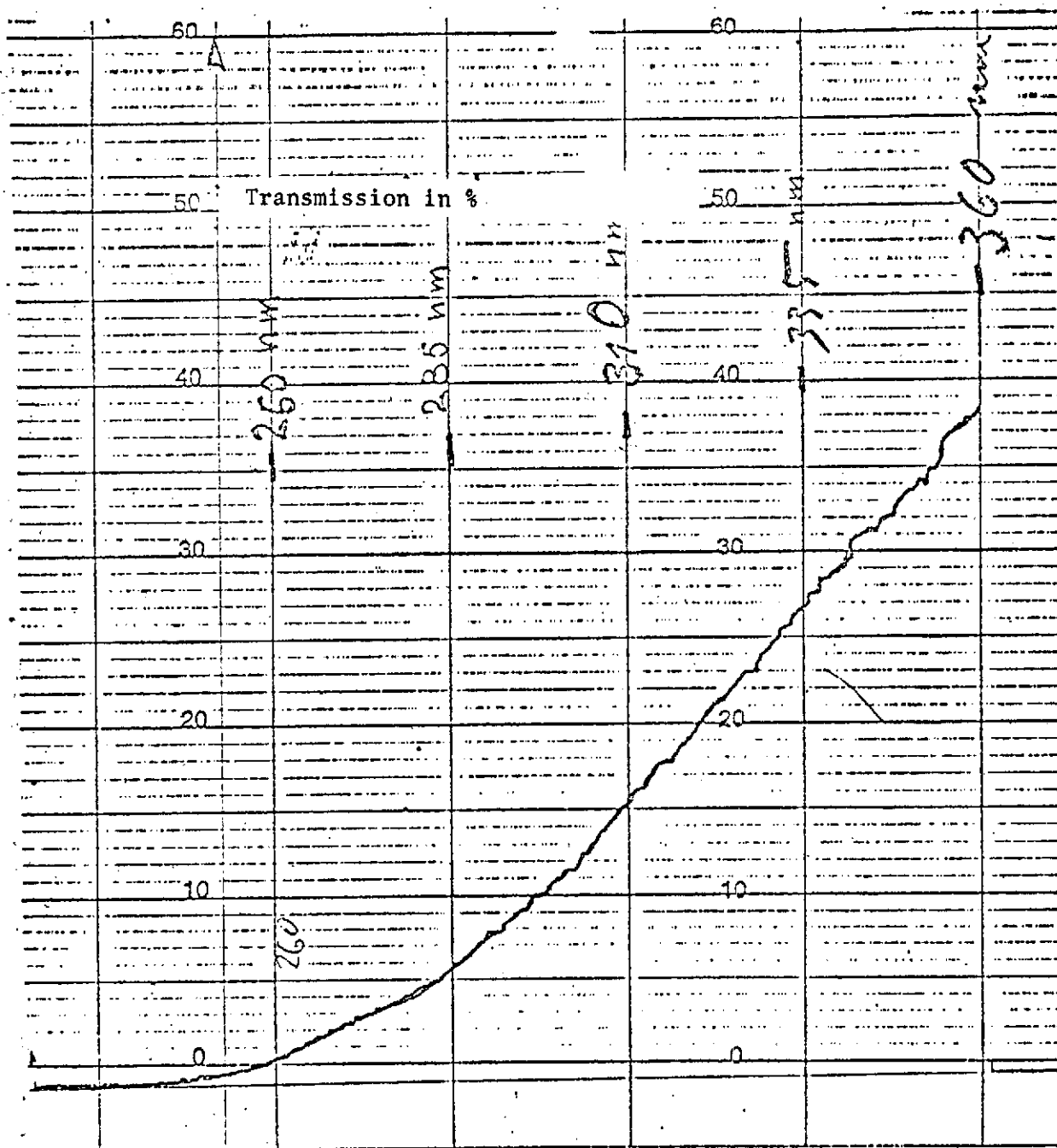


Figure 2. Transmission Curve in the Ultraviolet of a Silica Aerogel with a 0.16 Density and 4 mm Thickness.



Figure 3. Transmission Curve in Infrared of a Silica Aerogel with a 0.16 Density and 4 mm Thickness.

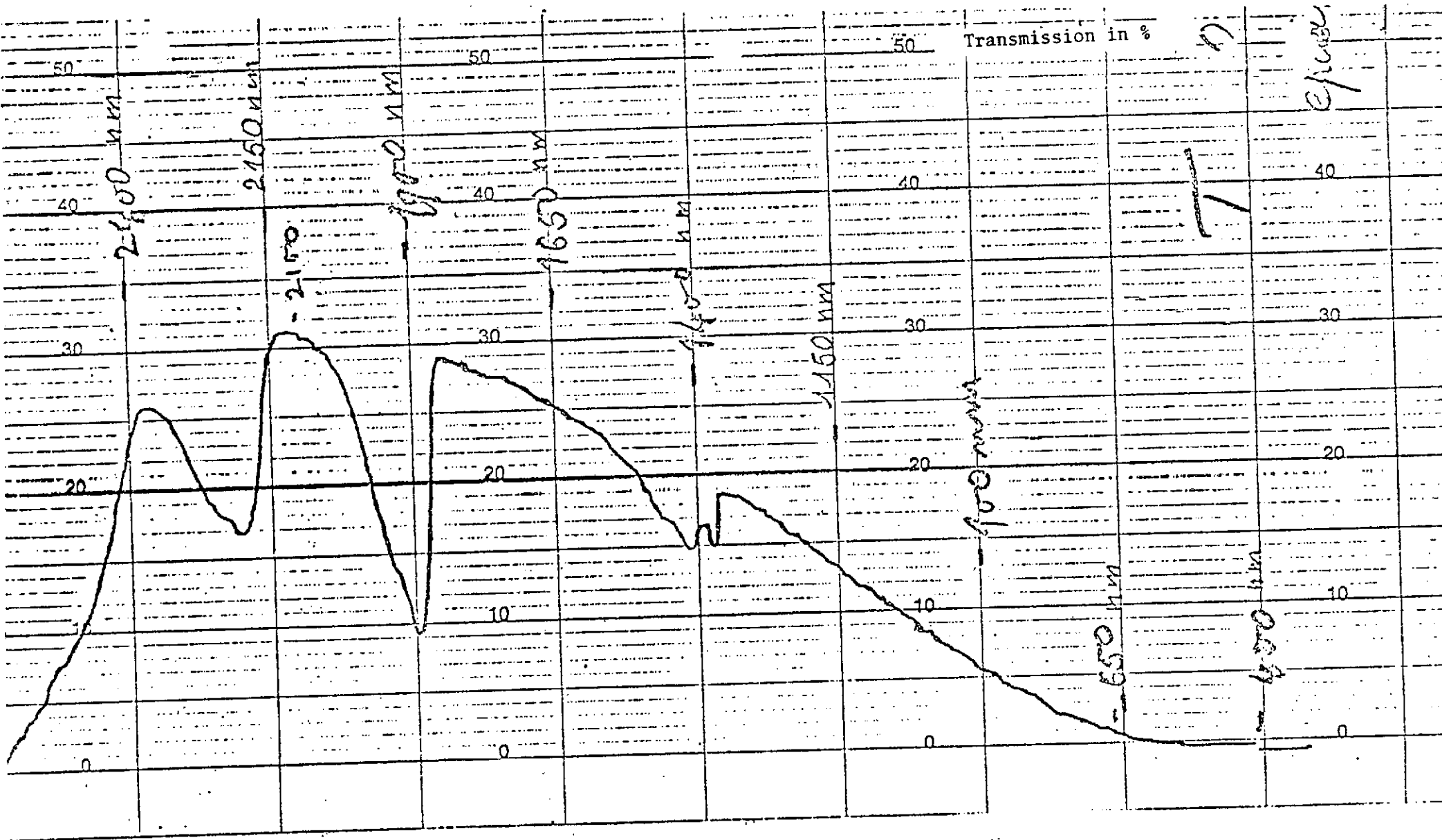


Figure 4. Transmission Curve in Visible and Infrared of a Compressed Silica Powder with a 1 Density and 2.4 mm Thickness.

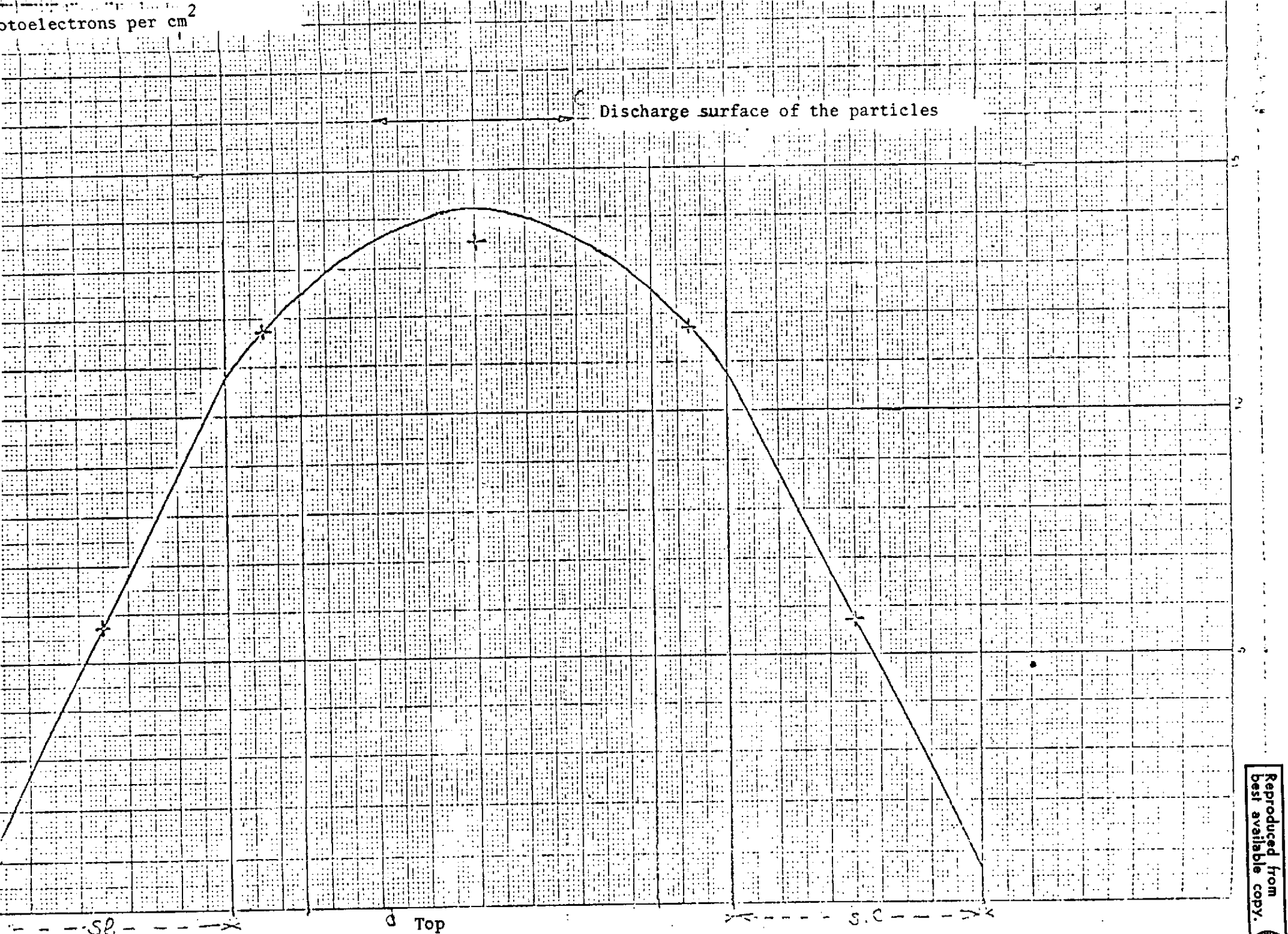


Figure 5. Distribution of the Photons at Discharge of an Aerogel with a Density of 0.3.

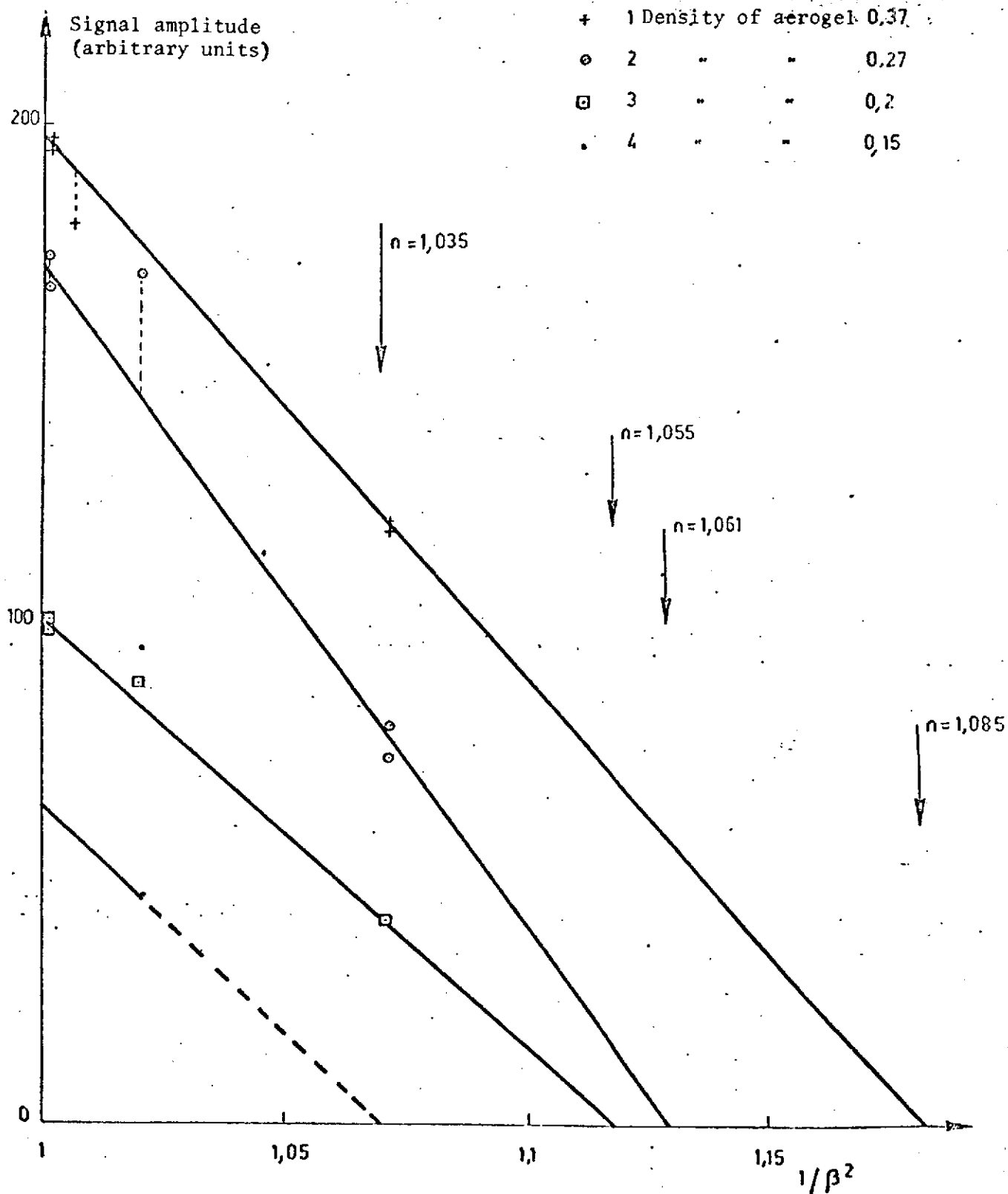


Figure 6. Response of a Silica Aerogel Passed Through by a Particle with Velocity βC as a Function of $1/\beta^2$. Commas indicate decimals.

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